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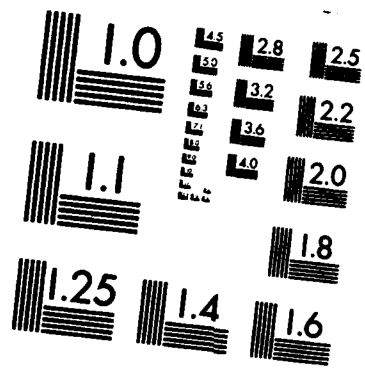

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ONR FINAL REPORT FOR CONTRACT N00014-81-K-0754

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The period of performance was August of 1981 through June of 1986. It is divided into two parts: one on nonlinear multiwave mixing, and one on theoretical studies relevant to free-electron lasers. Over the course of the years, the first part received more attention, and recently, a correspondingly larger portion of the funding. A description of the first part follows; the second part concludes this report.

PART I

Initially the contract was to investigate spectroscopic applications of four-wave mixing. This led naturally into laser and optical bistability instabilities, which are strongly affected by four-wave mixing, and to a quantum theory of multiwave mixing. This quantum theory shows how spontaneous emission affects and is affected by multiwave mixing both within and outside the optical cavities, and for one- and two-photon two-level media. The applications of this quantum theory include a more general theory of resonance fluorescence, build-up of optical instabilities from quantum noise, effects of quantum noise on pump/probe saturation spectroscopy, quantum effects on AM and FM modulation spectroscopy, and quantum effects on four-wave mixing. Although our contributions under ONR support to all of these areas have been significant, our work on quantum effects on four-wave mixing has received the greatest attention since it comprises the first nondegenerate theory of the generation of squeezed states in one- and two-photon two-level media. The area is very active, and three other groups have confirmed our results. We have benefited, in turn, from their observations and are currently pursuing studies of squeezed states under a new ONR contract.

In addition to the work on squeezed states, we have derived the two-photon resonance fluorescence spectrum for the first time. Dynamic Stark shifts can enter to modify the spectrum substantially from the one-photon case. Destructive interference can eliminate resonant Rayleigh scattering over some portions of the spectrum. We have also determined a quantum limit to the accuracy of modulation spectroscopy used, for example, by Bjorklund and by Hall. We have contributed several papers on the subject of laser and optical bistability instabilities, revealing in particular the central importance of population pulsations in causing these instabilities. We also developed the first theory that shows how the instabilities grow from spontaneous emission.

Over the years with ONR support, we have developed a semiclassical theory of two-photon multiwave mixing. Most recently we have generalized that theory to allow for an arbitrary amount of Doppler broadening. We find that the usual Doppler-free two-photon absorption only occurs when the Doppler width is large compared to the pump Rabi frequency, and we have studied the phase transition from homogeneous broadening to large Doppler broadening in detail.

The students that contributed to these efforts are: David A. Holm (PhD), Barbara A. Capron (PhD), Sami Hendow (PhD), Steve Stuut (MS), Lois Hoffer (MS, partially supported), and Shlomo Ovadia (PhD, partially supported).

The papers resulting from this ONR support in whole or in part are:

S. T. Hendow and M. Sargent III, "Effects of detuning on single-mode laser instabilities," Opt. Comm. 43, 59 (1982).

S. T. Hendow and M. Sargent III, "Role of population pulsations in laser and optical bistability instabilities," Opt. Comm. 40, 63 (1982).

M. Sargent III, M. S. Zubairy, and F. DeMartini, "Quantum theory of laser and optical bistability instabilities," Opt. Lett 8, 76 (1983).

S. Stuut and M. Sargent III, "Effects of Gaussian-beam averaging on phase conjugation and beat-frequency spectroscopy," J. Opt. Soc. Am. B 1, 95 (1984).

S. Ovadia and M. Sargent III, "Two-photon laser and optical bistability sidemode instabilities," Opt. Comm. 49, 447 (1984).

S. Ovadia, M. Sargent III, and S. T. Hendow, "Effects of dynamic Stark shifts on two-photon sidemode instabilities," Opt. Let. 10, 505 (1985).

S. T. Hendow and M. Sargent III, "Theory of single-mode laser instabilities," J. Opt. Soc. Am. B 2, 84 (1985).

M. Sargent III, S. Ovadia, and M. H. Lu, "Theory of two-photon multiwave mixing," Phys. Rev. A 32, 1596 (1985).

M. Sargent III, D. A. Holm, and M. S. Zubairy, "Quantum theory of multiwave mixing I. General formalism," Phy. Rev. A 31, 3112 (1985).

S. Stenholm, D. A. Holm, and M. Sargent III, "Quantum theory of multiwave mixing II. Operator approach," Phy. Rev. A 31, 3124 (1985).

D. A. Holm, M. Sargent III, and L. M. Hoffer, "Quantum theory of multiwave mixing III. Averages over inhomogeneous broadening, spatial hole burning, and Gaussian beams," Phys. Rev. A 32, 963 (1985).

D. A. Holm, M. Sargent III, and S. Stenholm, "Quantum theory of multiwave mixing IV. Effects of cavities on the spectrum of resonance fluorescence," J. Opt. Soc. Am. B 2, 1456 (1985).

D. A. Holm and M. Sargent III, "Theory of two-photon resonance fluorescence," Opt. Lett. 10, 405 (1985).

D. A. Holm and M. Sargent III, "Quantum theory of multiwave mixing V. Two-photon two-level model," Phy. Rev. A33, 1073 (1986).

D. A. Holm and M. Sargent III, "Quantum theory of multiwave mixing VI. Effects of quantum noise on modulation spectroscopy," J. Opt. Soc. Am. B3 732 (1986).

D. A. Holm and M. Sargent III, "Quantum theory of multiwave mixing VII. Connection to quantum Langevin theory," Phy. Rev. A 33 4001 (1986).

D. A. Holm, M. Sargent III, and B. A. Capron, "Generation of squeezed states by nondegenerate multiwave mixing in two-level media," Opt. Lett. 11, 443 (1986).

B. A. Capron, A. S. Marathay, M. Sargent III, "Theory of two-photon Doppler-broadened probe absorption," Opt. Lett. 11, 70 (1986).

B. A. Capron and M. Sargent III, "Effects of ionization and cascade decay on two-photon two-level interactions," accepted for publication in Phys. Rev. A.

B. A. Capron and M. Sargent III, "Theory of two-photon Doppler-free spectroscopy," accepted for publication in Phys. Rev. A.

In addition, approximately twelve talks were presented on these subjects at international meetings.

PART II

This part of the contract was to investigate properties of laser systems that have close analogies to the free-electron laser. The free-electron laser (FEL) has been of great interest as a tunable source of high-power radiation. FEL models are costly and difficult to compute, whereas analog cases in atomic systems are much easier. Atomic systems are easiest to use from, for example, the standpoint of developing scaling relations. Moreover they help sort out novel electromagnetic phenomena in the FEL from those attributable to using free electrons as a gain medium. This contract has concentrated on the consistency of the description of gain in FELs.

Analytical formulas describing energy extraction as a function of slippage have been developed and checked against numerical analysis. There is a single equation describing nonlinear behavior in all regimes of slippage that requires numerically generated values for the gain. Hence, while there are no contentious issue of gain in the FEL (there is a question of how to reconcile various approaches to computing the gain), it would be valuable to solve for the gain analytically, in which case we could dispense with the numerical calculation. The FEL at Los Alamos is expected to operate in the low slippage regime, whereas the Stanford device had high slippage. Timing of results is important to keep pace with experiment.

The problem of pulse amplification in the small signal regime of a laser amplifier is investigated theoretically. Analytical solutions to the Maxwell-Bloch equations are obtained. These reconcile the concept of laser gain needed to describe free-electron lasers (and synchronously pumped lasers in general) with earlier concepts that describe pulsed-laser amplifiers. We find the latter to be a transient of our solution while the former depends on its asymptotic features.

In summary we have shown how to extract general solutions to the linear differential equations that describe the process of gain in laser amplifiers. Because

of the unusual boundary conditions needed to achieve nonzero gains, the usual methods of analysis of linear partial differential equations are of little use. Nonetheless, we are able to write down solutions for arbitrary inputs in terms of expansions of supermodes. The coefficients of expansion, however, cannot be interpreted as probability amplitudes. We are uncertain whether the series possesses pointwise convergence, but it possesses an asymptotic convergence that makes the series a powerful tool for obtaining answers. We have shown that the conventional view of amplification as the inverse of absorption yields good approximation when restricted to transients.

Measurement of inverse absorption (g_{YL}) is usually done with a continuous wave (CW) field. In a laser, fields evolve to unknown configurations and theory is prejudicial insofar as it presupposes anything about them in advance. The single-mode CW laser that is paradigmatic in laser physics is special because the laser field conforms to and is often used as the field that measures gain. In this special case the YL gain is appropriate for quantum and semiclassical fields alike. It is not logical to expect that the results of the CW single-mode problem should generalize, especially with respect to quantum limits. A supermode analysis examines the gains that are appropriate for the set of all semiclassical pulses that circulate without change in the laser. The gains of supermodes are, in general, less than g_{YL}.

Whenever pulses with gains less than g_{YL} have been investigated, microscopic quantum fluctuations have been found to lead to macroscopic fluctuations in the classical fields (i.e., standard deviations are of the order of mean values). While we have not investigated the quantum problem, we find that our results confirm a testable result of previous numerical studies, namely that fluctuations should anticorrelate with pulse width. Observed high noise levels in synchronously pumped lasers are, however, often dismissed as artifacts of the same erratic pumps and jittering mirrors which prevent quantum-limited

performance in CW single-mode lasers. We suggest that it may be a waste of effort to try to achieve better performance through better pumps and better mirrors. The quantum limits of these devices should contain macroscopic fluctuations. We have made a specific predictions that may help to determine whether fundamental limits have already been reached.

Supermodes are interesting because their properties are almost independent of the model of the lasing medium. Synchronous pumping of Debye relaxations in a dye solvent should lead to results that are barely distinguishable from those of the FEL dye laser, if the solvent is pumped hard enough to reach the lasing threshold. Insensitivity to model comes from Maxwell's equations, most particularly from common boundary conditions that these equations impose on all models. We have designed a specific model that imposes the boundaries explicitly (the boundaries are usually implicit in numerical work). The fact that the boundaries yield modes that are non-self-adjoint and that are therefore difficult to handle mathematically does not justify falling back on the self-adjoint modes of free space as a substitute. The FEL has specifically refuted their use in practice and we have shown that they do not give generally sensible results in theory. Quantum problems in non-self-adjoint systems are currently of interest.[13] The methods for quantizing our model and for extracting specific predictions regarding fluctuations are known. The methods require only a prior solution of the semiclassical problem which we have given here.

The publications resulting from this ONR support in whole or in part include:

H. Al-Abawi, F. A. Hopf, G. T. Moore, and M. O. Scully, "Coherent transients in the free-electron laser: Laser lethargy and coherence brightening," Opt. Comm., 30, 1979.

A. L. Smirl, T. F. Boggess, and F. A. Hopf, "Generation of a forward-traveling phase-conjugate wave in Germanium," Opt. Comm. **34**, 463 (1980).

F. A. Hopf and J. Bergou, "Analytic solution of a laser amplifier with a delayed swept-gain boundary," Opt. Lett. 7, 411 (1982).

F. A. Hopf, "Steady-state pulses in a laser amplifier with delayed swept gain," Opt. Lett., 7, 605 (1982).

F. A. Hopf, "Delay effect in laser amplifiers," J. de Phys. **44**, C1-137 (1983).

F. A. Hopf and S. Stenholm, "Small-signal gain in lethargic and conventional laser amplifiers," Phys. Rev. A **28**, 1176 (1983).

F. A. Hopf, "Laser and Noise-driven operation in swept-gain lasers," Phys. Rev. A **27**, 2268 (1983).

F. A. Hopf, "Influence of slippage parameter on swept-gain amplifiers," Phys. Rev. A, **30**, 6 (1984).

F. A. Hopf, J. Bergou, and S. Varro, "Transient and asymptotic small signal gain in laser amplifiers," to be published.

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